

perfect material, are thus confirmed in the most gratifying manner. Many problems yet remain to be solved, however, especially the osteology of the line leading directly into the modern horse. Explorations will therefore be continued, especially the search for the skeleton of *Protohippus*, with a view to ascertaining whether this is or is not one of the direct ancestors of *Equus caballus*.

American Museum of Natural History,
New York, April 7, 1904.

RADIUM IN AN AMERICAN ORE.

BY ALEXANDER H. PHILLIPS.

(Read April 8, 1904.)

The work which I have accomplished in the separation of radium, or more exactly the concentration of radium in barium salts, has been carried on entirely with the mineral carnotite.

Carnotite is comparatively a new mineral, having been described by Friedel and Cumenge in July, 1899, and for this reason it is not found in most books on mineralogy, and is therefore but little known to the general prospector. It was first discovered in the western part of Colorado, and occurs in Montrose, San Miguel and Mesa counties of that State and the adjacent counties of Utah.

The theoretical percentage composition as given by Friedel and Cumenge is:

UO ₃	63.54%
V ₂ O ₅	20.12 "
K ₂ O	10.37 "
H ₂ O	5.95 "

Results very close to these were obtained in the actual analyses. The mineral formula is given as 2UO₃, V₂O₅, K₂O, 3H₂O, or a uranyl potassium vanadate with three molecules of water of crystallization. Hillebrand, after a series of analyses, disputes this composition, and holds that the mineral is probably a mixture to which the above simple formula is not applicable.

Carnotite is a light canary-colored powder disseminated through a fine-grain sandstone. It is easily soluble in acids, and is treated in this way for the commercial production of uranium salts.

In October, 1902, I received twenty-five pounds of this ore from Richardson, Utah. Carnotite occurs here in the usual way; the ore being rather a lean one, no specimens of which carries more than 10% of the mineral carnotite, while the average is greatly below this amount. The percentage content of uranium and vanadium in this ore was not determined, which is to be regretted.

The radio-activity compared to uranium, as determined by G. B. Pegram, of Columbia University, was .40.

After a series of experiments upon a one-pound sample, the remainder of the twenty-five pounds was treated as follows: It was first leached with hydrochloric acid to remove most of the soluble salts; as radium salts are isomorphous with barium salts, and agree very closely in their chemical properties and solubilities with the similar barium salts, it was thought that strong nitric acid would dissolve the small amount of barium the ore contained and also the radium with it.

After the hydrochloric acid treatment the insoluble residue was treated with concentrated nitric; these two acid solutions were concentrated to small volume. Upon testing them for barium it was found that the precipitate would be small, so it was thought advisable to add a small amount of barium chloride, which would act as a carrier and help in the separation of the small amount of radium present; sulphuric acid was then added; the solutions diluted and allowed to stand several days to settle; the clear solutions siphoned off. The resulting sulphates after washing were fused with alkali carbonates. The melt dissolved in water, the insoluble carbonates after washing free of sulphuric acid were dissolved in hydrochloric acid. Hydrogen sulphide was then passed through the solution to free it of lead. From this solution free of lead the barium was precipitated as a carbonate, and dissolved in the least possible quantity of hydrochloric acid, when the barium and radium will be in the form of chlorides and are in a condition to concentrate the radium by fractional crystallization. This solution of chlorides was allowed to evaporate slowly until about one-half of the contained salts had separated as crystals, when the crystals were removed, redissolved, the solution again allowed to evaporate slowly until one-half had separated, and so on for a third time, when there was obtained as a final product a little less than one-half gramme of chlorides, the activity of which compared to uranyl nitrate was about 1500.

The residual chlorides amounting to two grammes were recovered, and proved to be quite active also; their activity as compared to uranium was measured by Pegram as 365.

The radio-activity of the final product could be increased by several fractional crystallizations, when a specimen much less in weight, but more active, would be obtained.

The radio-activity of the specimen as obtained was deemed sufficiently high to indicate that radium could be produced in quantity from carnotite, at least from this locality, as twenty-five pounds of rather a lean ore had been used. Had a ton been worked over in the same way it would yield a gramme of chlorides of 60,000 radio-activity as compared to uranium. This specimen was separated in November, 1902, and is as active now as then.

This establishes without doubt the fact that radium salts are dissolved in the acid with which the uranium minerals are treated in the commercial preparation of uranium salts. In the crystallization of these salts the radium would be carried in connection with the uranium, as it is in the natural formation of the uranium minerals. This would explain to a certain extent the variable activity of uranium salts, as their activity is not proportional generally to the uranium which they contain.

Since the separation of this first sample of radium from carnotite, I have received specimens of the mineral from other localities, all of which are active, their activity depending upon the amount of carnotite in the ore. One specimen of quite pure mineral gave an activity of 4, the highest observed in the crude ore.

A short time ago 3.5 kilos (about eight pounds) were obtained from Montrose County, Colo., selected specimens of which were exposed, in the ordinary way, in making X-ray negatives, with very satisfactory results.

These photographs show very clearly the bands of active carnotite separated by the inactive matrix.

The plates used were Carbutt's B, and exposed to the action of the mineral for sixty hours. With more sensitive plates the same effect could be obtained in much shorter time.

I am at present at work upon the separation of the uranium, vanadium, and radium salts from these eight pounds of ore as an exhibit at the St. Louis Fair. This work is as yet not completed.

After pulverizing and thoroughly mixing, its radio-activity was measured as 1.71, compared to uranyl nitrate. It was then treated

with dilute acids, as it was intended in this case to boil the insoluble residue in a concentrated solution of sodium carbonate, to extract the last of the radium. After the acid treatment the residue from the 3500 gms. originally taken weighed 2200 gms. and gave an activity of 1.40. The solution contained 1300 gms. of the amount taken.

If the activity is calculated to gramme units compared to uranyl nitrate the 3500 gms., activity $1.71 = 5985$ units.

Residue insoluble in dilute acids, 2200 gms., activity $1.40 = 3080$ units. The solution contained, therefore, 2905 units, considerable of which would be due to the uranium dissolved and emanation from the radium. The dilute acid extracted nearly all of the uranium, but there was still some found in the residue. This ore contained considerable barium and a large amount of calcium. No barium was therefore added to the solutions, as before. After precipitation of the sulphates, and the separation of barium from other bases, 3.8 gms. of barium carbonate were obtained, which gave an activity of 35.8, or 135 units compared to uranium nitrate. This activity was measured upon the same day (*i.e.*, yesterday) that the barium salt was separated from the other bases; it may be expected that its activity will increase, from the accumulation of the emanation; this increase may in some cases amount to several times the original activity of the compound when first prepared.

This demonstrates that dilute acids, while they dissolve considerable of the barium salts present, the greater proportion of the radium is still left in the residue; but even this amount, which is small compared to that left in the residue, as indicated by the radio-activity of the residue, if calculated upon the basis of a ton, would yield a gramme of chlorides of 11,300 activity as compared to uranium.

These facts prove beyond question that carnotite will become a commercial source of radium.

Princeton University, March 7, 1904.